

The Effectiveness of Hydrogen Water Chemistry in a Boiling Water Reactor during Power Coastdown Operations

Tsung-Kuang Yeh¹, Mei-Ya Wang², Charles F. Chu³, Ching Chang³

¹Department of Engineering and System Science, National Tsing Hua University, Taiwan

²Nuclear Science and Technology Development Center, National Tsing Hua University, Taiwan

³ Nuclear Generation Department, Taiwan Power Company, Taiwan

¹tkyeh@mx.nthu.edu.tw; ²meywang@mx.nthu.edu.tw; ³u810111@taipower.com.tw; ³u808675@taipower.com.tw

Abstract

A theoretical model was adapted to evaluate the impact of power coastdown on the water chemistry of a commercial boiling water reactor (BWR) in this work. In principle, the power density of a nuclear reactor upon a power level decrease would immediately be lowered, followed by water chemistry variations due to reduced radiolysis of water and extended coolant residence times in the core and near-core regions. It is currently a common practice for a commercial BWR to adopt hydrogen water chemistry (HWC) for corrosion mitigation. The optimal feedwater hydrogen concentration may be different after a power coastdown is implemented in a BWR. A computer code DEMACE was used in the current study to investigate the impact of power coastdown on major radiolytic species concentrations and electrochemical corrosion potential (ECP) behavior of components in the primary coolant circuit of a domestic reactor operating under either normal water chemistry or HWC. Our analyses indicated that under a rated core flow rate the chemical species concentrations and the ECP did not vary monotonously with decreases in reactor power level at a fixed feedwater hydrogen concentration. In particular, ECP variations basically followed the patterns of hydrogen peroxide in the select regions and exhibited high values at power level of 80% for Reactor X.

Keywords

BWR; Water Chemistry; Power Coastdown; Hydrogen Water Chemistry; IGSCC

Introduction

Current light water reactors (LWRs) produce electric energy at their rated power level until the reactivity-limited burnup is reached. When all control rods are fully withdrawn and core flow is at or near the rated value to generate maximum power in an LWR near the end of its fuel cycle, a coastdown operation (i.e. gradually reduced power) may commence. A typical

coastdown duration may last from a week to a month with a terminal operating power level at 80% to less than 100% of the rated value. The operating power would decrease one percent in about every two days. In a similar manner, a boiling water reactor (BWR) may undergo a power coastdown operation near the end of a fuel cycle when the fuel in the core is sufficiently depleted earlier than scheduled and the rated power can no longer be maintained. Upon a power coastdown, the power density and coolant flow velocity in the reactor core would accordingly change, followed by water chemistry variations due to reduced radiolysis of water and extended coolant residence times in the near-core regions.

Intergranular stress corrosion cracking (IGSCC) on stainless steel and nickel-base alloy components has been a major material degradation issue for decades for all BWRs around the world. The IGSCC susceptibility of structural materials in a BWR is primarily dominated by the physical and chemical properties of the materials, tensile stresses, and the primary coolant chemistry. For mitigating IGSCC in an operating BWR, improvement on the primary coolant chemistry by hydrogen water chemistry (HWC) has been adopted worldwide [1]. The HWC technology is deemed effective when the electrochemical corrosion potential (ECP) in the primary coolant circuit (PCC) of a BWR at a specific feedwater hydrogen concentration ($[H_2]_{FW}$) is reduced to below $-0.23 V_{SHE}$ that signifies immunity of structural components to IGSCC. The U.S. Nuclear Regulatory Commission has recognized an ECP value of $-0.23 V_{SHE}$ to be the critical corrosion potential (E_{crit}) of sensitized Type 304 stainless steels [electrochemical potentiodynamic reactivation (EPR) = $15 C/cm^2$] in

typical BWR environments (i.e., a coolant temperature > 270 oC and coolant conductivity near $0.3 \mu\text{S}/\text{cm}$). If a BWR is operating at full power under HWC, the optimal $[\text{H}_2]\text{FW}$ that varies from plant to plant would usually be set at a constant value. In theory, a reduced power level as in the case of a coastdown operation would induce different water chemistry in the reactor coolant, possibly leading to a different $[\text{H}_2]\text{FW}$ demand to maintain the corrosion potential below Ecr.

Upon a power coastdown, the power density of the BWR would decrease, and the mass flow rate in the PCC would remain at the rated core flow rate. The lower power density in the reactor core will lead to a less production of steam in the core fuel channels, and this will in turn result in lower linear liquid and steam velocities in the boiling core region and lengthen the total residence time of the coolant in the core region. The combined effect of lower neutron and gamma photon dose rates and an extended coolant residence time in the reactor core complicates the water chemistry in the PCC of a BWR due to less radiolysis but an enhanced irradiated time of the primary coolant.

The amounts of major redox species (i.e. hydrogen, oxygen, and hydrogen peroxide) present in the reactor coolant determine through the mixed potential effect the ECP in the PCC of a BWR. The concentrations of these redox species, however, are directly related to the degree of water radiolysis, which is dominated by the power density of the reactor and by the coolant residence time in the reactor core. Changes in power density (i.e. neutron and gamma photon dose rates) and coolant flow velocity in the reactor core would lead to concentration variations of the major redox species [2-6]. Accordingly, the required $[\text{H}_2]\text{FW}$ s at a reduced power level for the HWC technology to take effect on IGSCC mitigation may thus be different. In the meantime, it is also anticipated that the impact of a reduced power level on the effectiveness of HWC may also be different in different regions of a PCC in a BWR.

The concentrations of radiolysis products and the ECP values differ significantly in the PCC of a BWR. It is very difficult to actually measure the water chemistry data directly at various locations of an actual reactor. Accordingly, the impact of power coastdown on the water chemistry of a BWR operating under HWC can only be evaluated theoretically through computer modeling. A deterministic model by the name of

DEMACE was developed in the past for analyzing water chemistry variation and corrosion behavior of metallic materials in the PCC of a BWR [4-6]. In the current study, a local BWR designated as Reactor X was selected for demonstrating the impact of different power coastdown conditions on the major redox species concentrations and on the ECP behavior of structural components in the PCC. Reactor X is a BWR/4 type reactor with rated thermal power of 1775 MW. It is currently operating under HWC at a 0.5 parts per million (ppm) $[\text{H}_2]\text{FW}$. Numerical simulation for Reactor X was carried out for $[\text{H}_2]\text{FW}$ ranging from 0.0 to 2.0 ppm and for power levels ranging from 80% to 100%. Variations in oxygen (O_2), hydrogen (H_2), and hydrogen peroxide (H_2O_2) concentrations and in ECP at four selected locations in the PCC of Reactor X were analyzed. The impact of power coastdown on the corrosion mitigation effectiveness of HWC is discussed. Optimal HWC operating condition for Reactor X under power coastdown operation is then proposed.

Modeling Approaches

For simplicity in modeling, the entire PCC of a BWR was divided into 12 regions in the DEMACE computer model, as shown in Figure 1. The DEMACE computer code consists of a radiolysis model for calculating chemical species concentration, a mixed potential model for calculating ECP, and a coupled environment fracture model for calculating the crack growth rate. More detailed descriptions about these

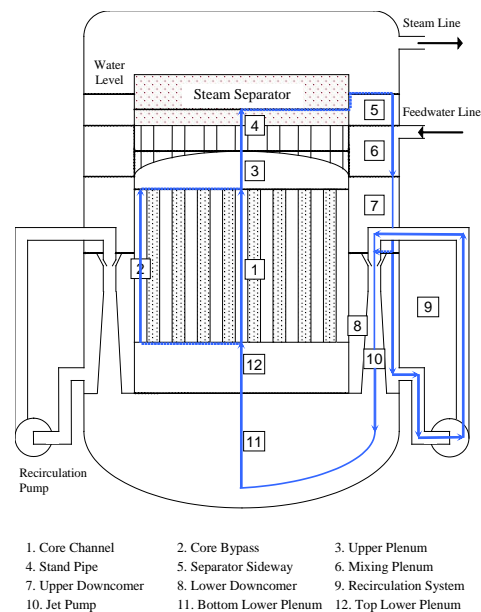


FIG. 1 CONCEPTUAL CONFIGURATION OF A TYPICAL BWR PRIMARY COOLANT CIRCUIT

three models can be found in the literature [4-8]. Our earlier work dealt with the impact of power uprate on water chemistry in Reactor X and showed that the degree and pattern of the impact actually varied from location to location [9-10]. In the current study, we intend to find out if a similar outcome would appear during power coastdown operation.

Power Coastdown Simulations

During the course of a coastdown operation in a BWR, the core flow rate in the BWR is usually maintained at the rated value in order to generate the greatest power. Therefore, rated core flow rate was used for Reactor X during the simulation. The dose rates of neutron and gamma photon were assumed to vary linearly as the reactor power level decreased. The linear flow velocities of the liquid coolant in the upper plenum, stand pipe, and separator sideways regions were recalculated based on the steam quality produced and on the liquid left in the reactor core at different power levels. The steam quality (X) is defined as the mass fraction of vapor in the mixture of liquid and vapor and can be expressed by

$$X = \frac{H - H_f + H_i}{H_{fg}}, \quad (1)$$

where H is the enthalpy of the mixture at the core outlet, H_f is the enthalpy of the saturated liquid at the system pressure, H_i is the enthalpy of the liquid at the core inlet, and H_{fg} is the enthalpy of vaporization at the system pressure. H is a function of total produced power and core flow rate and may be expressed as

$$H = \frac{F_c Q_c}{m}, \quad (2)$$

where F_c is the hot channel factor in the core, Q_c is the total produced power in the core, and m is the core flow rate. As the reactor power level decreases at a fixed pressure and a fixed core flow rate, H would accordingly decrease and lead to a decrease in X .

The decreases in steam quality as a function of reactor power level were calculated using the ZEBRA computer code [11], and the results are shown in Figure 2. For the core channel region, the variation in coolant flow velocity along the core channel was dominated by the steam quality (and hence the void fraction), and the liquid flow velocity became lower as the steam quality decreased in this region at a fixed power level. The foregoing changes at different power levels in radiation dose rates and in coolant flow

velocity were expected to influence the steady-state water chemistry of the reactor since the degree of radiolysis and the time for the coolant to undergo radiolysis were altered.

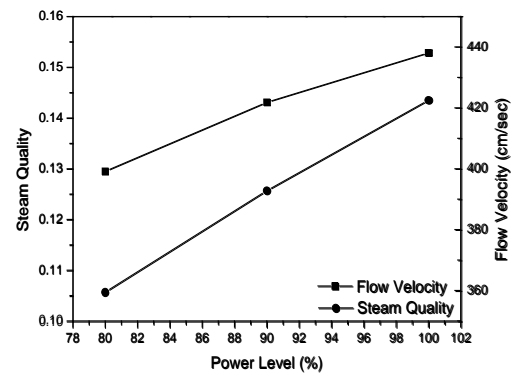


FIG. 2 STEAM QUALITY AND LIQUID COOLANT FLOW VELOCITY AS A FUNCTION OF POWER LEVEL AT THE CORE EXIT OF REACTOR X OPERATING AT THE RATED MASS FLOW RATE.

Modeling Procedures

Before the actual modeling work on Reactor X under various power coastdown conditions commenced, the DEMACE computer model was deliberately calibrated by fitting the predicted O_2 concentration at the recirculation system outlet of this BWR with measured data, obtained from the results of the HWC ramping test conducted in 2006. The operating conditions of Reactor X in 2006 were used in the model calculation during the calibration process. By adjusting the gas transfer coefficients in the core boiling channels of Reactor X, a routinely taken technique [3-5], and by applying the least square fitting algorithm, we were able to obtain predicted results that are in reasonably good agreement with the measured plant data.

The power coastdown condition considered in this study was 80% of the rated power level. The rated power condition was also taken into account for comparison purpose. At a specific power level, the coolant flow velocities in the core and other near-core regions were recalculated based upon the steam quality produced in the core fuel channels. The dose rates of neutron and gamma photon in Reactor X at rated power were calculated using an independent computer code by the name of DRONG [12]. The dose rates neutron and gamma photon during coastdown operations were then assumed to vary linearly with the power level. Furthermore, a different but lower power level at the rated coolant flow rate during a coastdown operation would enable the calculation of steam quality and void fraction in the core region, and

this part of the study was done using the ZEBRA computer code. Changes in void fraction in the core boiling region of a BWR are expected to have an important impact on the effectiveness of HWC since gas transfer rates of O_2 and H_2 between the steam and liquid phases in this region are altered accordingly. After the foregoing parameters were derived, water chemistry and ECP modeling was then carried out for the entire PCC of Reactor X with $[H_2]FW$ s ranging from 0.0 to 2.0 ppm. To avoid lengthy discussion, only four particular locations of major IGSCC concerns in Reactor X were selected to demonstrate the impact of various power coastdown conditions on the effectiveness of HWC. The four specific locations of major IGSCC concerns selected were the outlet of the upper plenum region (Location 3), the outlet of the upper downcomer region (Location 7), the outlet of the recirculation system (Location 9), and the outlet of the bottom lower plenum region (Location 11) which approximately represent the top guide, the belt area of the core shroud and jet pump risers, the recirculation piping, and the core shroud base and support, respectively. These locations have a long history of IGSCC incidents as reported in the literature [13-15].

Results and Discussion

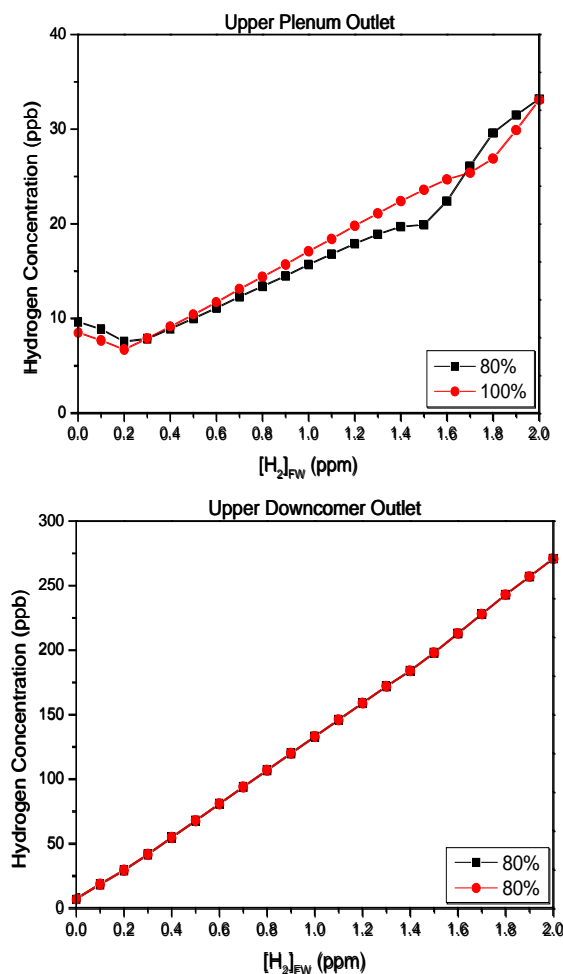
Simulation results are categorized into two portions showing variations in the concentrations of major redox species and in the ECP as a function of $[H_2]FW$ at the four selected locations.

Species Concentrations

Although there are a total of eleven radiolysis species taken into account in the DEMACE model, only three major redox species with relatively significant concentrations are used in the calculation of ECP. Details on the full set of chemical reactions among these species and their respective rate constants can be found elsewhere [4]. Among the eleven radiolysis species (e^- , H , H^+ , OH , OH^- , H_2O_2 , HO_2 , HO_2^- , O_2 , O_2^- , and H_2), H_2 and H_2O_2 are directly produced during the course of water radiolysis, and O_2 is then produced through the decomposition of H_2O_2 , not a direct radiolysis product. These three species actually dominate the magnitude of ECP because the concentrations of the other redox species (e.g. OH and HO_2) are comparatively small and can thus be neglected. The concentration variations of the three major species as a function of $[H_2]FW$ at the designated locations of Reactor X are of great importance.

Concentrations of H_2 :

The predicted H_2 concentrations ($[H_2]$ s) in the reactor coolant are shown in Figure 3 for Reactor X. It was observed that at a fixed power level the $[H_2]$ monotonically increased with increasing $[H_2]FW$ up to 2.0 ppm at the outlets of the upper downcomer, the recirculation system, and the bottom lower plenum. However, the $[H_2]$ was relatively low at the upper plenum outlet, which was resulted from the gas stripping process in the core boiling region, although it still increased with increasing $[H_2]FW$ up to a maximum of more than 30 parts per billion (ppb) at 2.0 ppm $[H_2]FW$. Under power coastdown conditions, it seemed that a lower power level would lead to a lower $[H_2]$ in the reactor coolant at all four locations. However, there was one exception at the upper plenum outlet of Reactor X, and the $[H_2]$ at 100% power was lower than that at 80% power at $[H_2]FW$ s of less than 0.3 ppm and higher than 1.6 ppm. For Reactor X, no significant differences in $[H_2]$ were observed for both power levels at low $[H_2]FW$ s (i.e. <0.6 ppm).



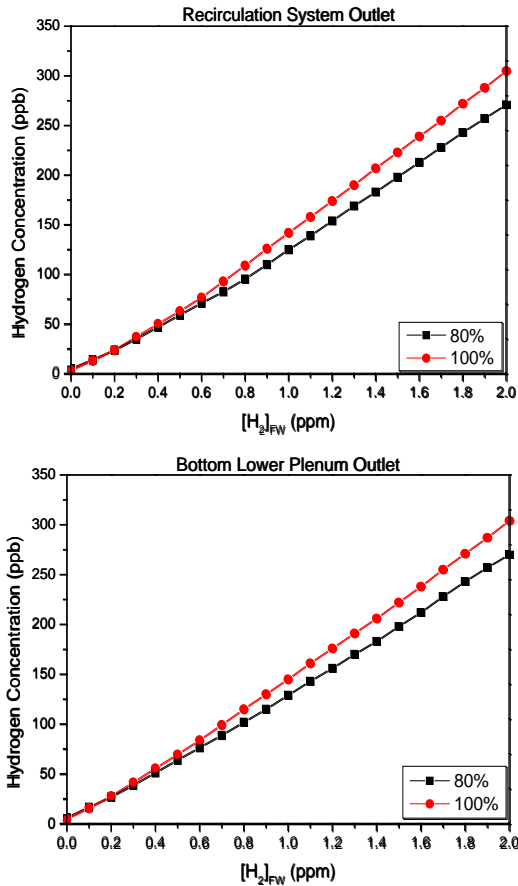


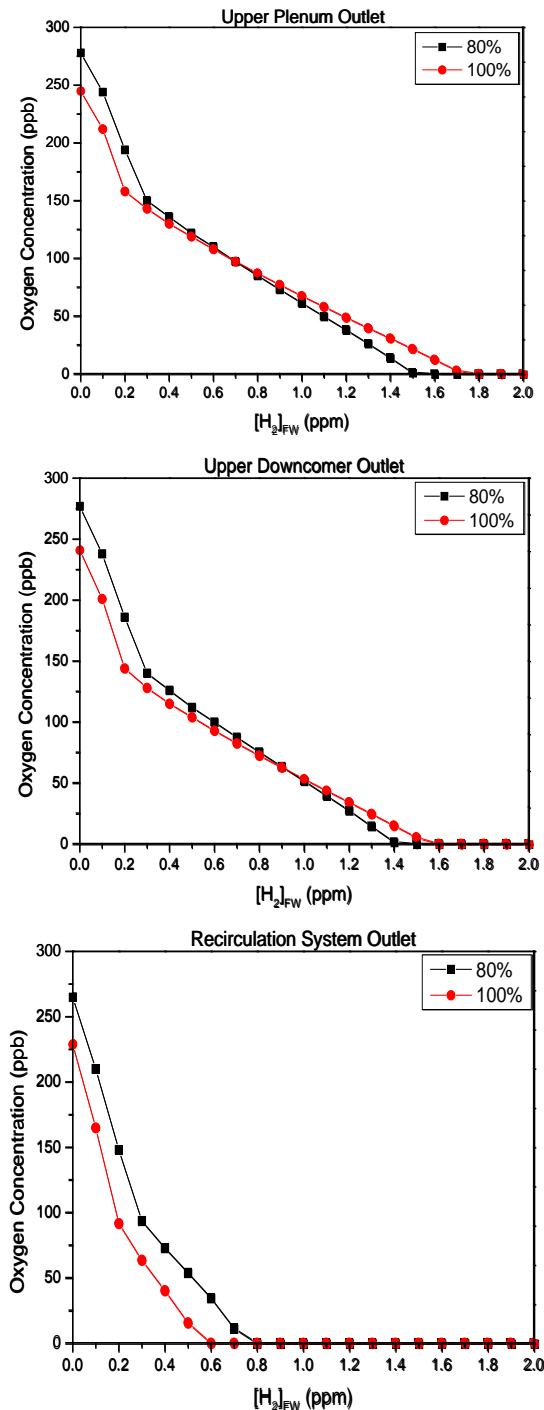
FIG. 3 VARIATIONS IN $[H_2]$ AS A FUNCTION OF $[H_2]_{FW}$ AT FOUR SELECTED LOCATIONS OF REACTOR X WITH OPERATING POWER LEVELS OF 80% AND 100% OF THE RATED POWER.

Concentrations of O_2 :

Variations in O_2 concentration ($[O_2]$) at the selected locations of Reactor X are shown in Figure 4. In general, a higher $[H_2]$ FW would lead to lower predicted $[O_2]$ s at all four locations no matter what the power level was. Among these locations, the $[O_2]$ at the recirculation system outlet was reduced most effectively under HWC, with a 0.8 ppm $[H_2]$ FW being sufficient to scavenge all dissolved O_2 in the coolant. In the meantime, the dissolved O_2 in the coolant at either one of the selected power levels was not fully depleted until the $[H_2]$ FW reached 1.6 ppm and 1.4 ppm at the upper downcomer outlet and at the bottom lower plenum outlet, respectively. While 1.6 ppm of $[H_2]$ FW was necessary to reduce the $[O_2]$ to zero at 80% power level at the upper plenum outlet, the $[O_2]$ went down to zero at 1.8 ppm of $[H_2]$ FW at 100% power level. It was clearly demonstrated that the effectiveness of HWC on reducing the $[O_2]$ in a BWR might vary from region to region along the PCC due to different degrees of radiolysis in these regions.

In view of the effect of power level, we found that

decreases in power level generally led to increases in $[O_2]$ at all selected locations of Reactor X. The differences in $[O_2]$ could be as much as more than 40 ppb between the power levels of 80% and 100% at certain $[H_2]$ FWs. Relatively large differences in $[O_2]$ were distinctly observed at low $[H_2]$ FWs (i.e. <0.7 ppm or smaller). It was therefore foreseen that a 20% power coastdown operation causing undesirably higher $[O_2]$ s at low $[H_2]$ FWs would eventually influence the effectiveness of HWC on corrosion mitigation in Reactor X, and it was indeed proven so based upon the predicted ECP results discussed later.



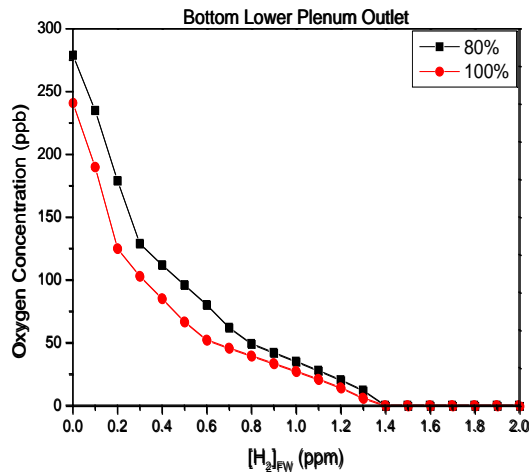
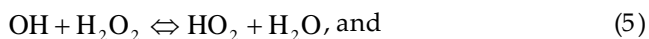


FIG. 4 VARIATIONS IN [O₂] AS A FUNCTION OF [H₂]FW AT FOUR SELECTED LOCATIONS OF REACTOR X WITH OPERATING POWER LEVELS OF 80% AND 100% OF THE RATED POWER.

Concentrations of H₂O₂:

The other important oxidizing species in the BWR coolant is H₂O₂, a direct radiolysis product. Figure 5 exhibits variations in H₂O₂ concentration ([H₂O₂]) as a function of [H₂]FW at the selected locations of Reactor X. In contrast to the [O₂], the [H₂O₂] did not decrease monotonically as the [H₂]FW increased. Except for the upper plenum outlet, the [H₂O₂]s were predicted to first increase and decrease later on with increasing [H₂]FW. Four chemical reactions were speculated to be responsible for this unique outcome, and they are



Equation (3) with the greatest rate constant of all dominated the concentrations of species H and OH. When a mild amount of H₂ was added into the reactor coolant, Equation (4) started taking effect and more H was produced. More H then led to more consumption of OH via Equation (3), which in turn led to less consumption of H₂O₂ via Equation (5). Although some H₂O₂ could have reacted with H via Equation (6), the decrease in [H₂O₂] was comparatively less significant. Once the [H₂]FW increased to an effectively high level, the concentration of H generated via Equation (4) became vital and Equation (6) started to dominate. The [H₂O₂] then promptly decreased via both Equations (5) and (6) as seen in Figure 5. Since H₂O₂ is a highly oxidizing species and a few ppb H₂O₂ could render the ECP well above the Ecrit, it is essential to eliminate all

H₂O₂ in the coolant to achieve considerably low ECPs in the PCC of a BWR. Due to differences in degree of radiolysis, the [H₂O₂] at various locations responded to the [H₂]FW quite differently at selected power levels.

For a fixed power level, the [H₂O₂] at the upper plenum outlet of Reactor X steadily decreased as the [H₂]FW increased from 0 to 0.2 ppm and then slowly decreased with increasing [H₂]FW. The [H₂O₂] at this location would be more than 14 ppb at 1.7 ppm [H₂]FW at 100% power level, but it became zero at the same [H₂]FW at the 80% power level. In view of the impact of power level, we found that the [H₂O₂] generally increased with decreasing power level at all [H₂]FWs. With increasing [H₂]FW, the [H₂O₂] reduced with decreasing power level. However, at the 80% power level, the [H₂O₂] was initially higher than that at the rated power at low [H₂]FWs (i.e. <0.3 ppm) and then became comparatively lower at high [H₂]FWs. This unique phenomenon highlighted our argument that a combined effect of reduced radiolysis and prolonged coolant residence time in the reactor core upon a coastdown operation could arbitrarily alter the water chemistry in the PCC of a BWR.

At the upper downcomer outlets, the response of [H₂O₂] to [H₂]FW was quite different from that at the upper plenum outlet. Variations in power level did not cause any significant changes in [H₂O₂] for Reactor X operating under normal water chemistry. It was noted that a 1.5 ppm [H₂]FW was required to scavenge all H₂O₂ in the coolant at this location at the power levels of 80%, but the [H₂O₂] remained more than 19 ppb at 100% power at the same [H₂]FW. In general, a lower power level would render a more effective reduction of H₂O₂ no matter what the [H₂]FW was for Reactor X.

In contrast to the outcome at the previous two locations, a higher power level would instead lead to a more effective reduction in [H₂O₂] at the recirculation system outlet. A coastdown operation of 80% power would alter the HWC effectiveness on scavenging H₂O₂ from 0.6 ppm to 0.8 ppm.

Variations in power level did cause any significant changes in [H₂O₂] at most [H₂]FWs (<1.3 ppm) at the bottom lower plenum outlet of Reactor X. The required [H₂]FWs to effectively eliminate H₂O₂ in the coolant were predicted to be 1.4 ppm at all selected power levels. Inconsistent trends in [H₂O₂] at various [H₂]FWs resulted from changes in power level highlighted the effect of the foregoing competing process of radiolysis and residence time.

Summarizing the concentration results from the two oxidizing species, we found that for Reactor X a 5% power reduction could promote a more oxidizing coolant environment for the structural components positioned at the selected locations. Increases in the concentrations of H_2O_2 and O_2 at a particular coastdown level would pose a significant impact on ECP according to the mixed potential model [7]. Therefore, a similar outcome is expected to appear in ECP response to power coastdown operation.

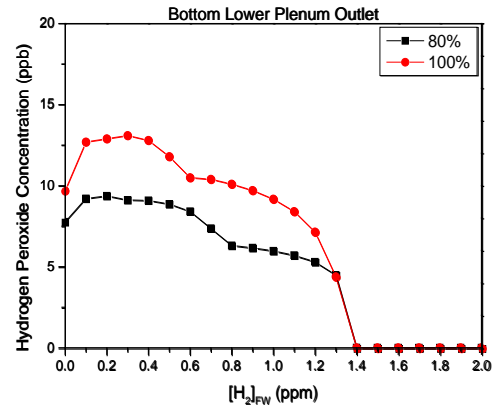
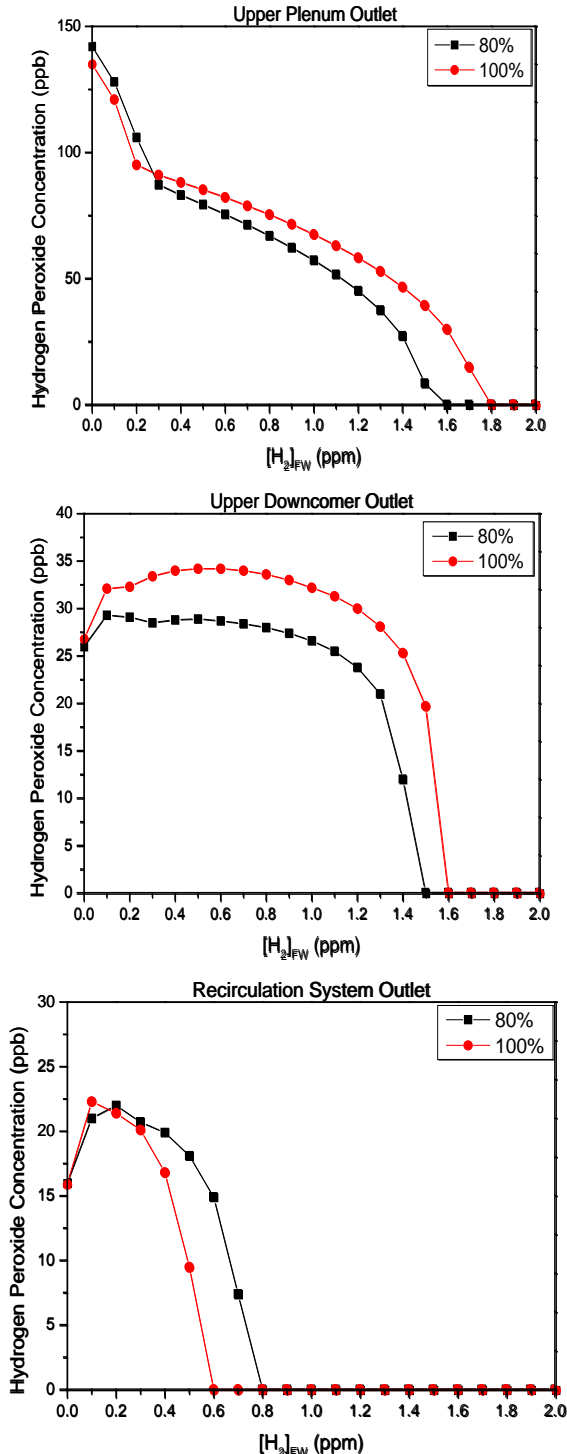


FIG. 5 VARIATIONS IN $[\text{H}_2\text{O}_2]$ AS A FUNCTION OF $[\text{H}_2]\text{FW}$ AT FOUR SELECTED LOCATIONS OF REACTOR X WITH OPERATING POWER LEVELS OF 80% AND 100% OF THE RATED POWER.

ECP

In the current study, the well recognized ECP of $-0.23 \text{ V}_{\text{SHE}}$ was selected as the Ecrit of highly sensitized Type 304 stainless steels in typical BWR environments.

Based upon this criterion, a component is considered protected from IGSCC and the HWC technique is considered effective if the predicted ECP is below the Ecrit. Figures 6 shows variations in ECP as a function of $[\text{H}_2]\text{FW}$ and power level at the four selected locations of Reactor X.

At the upper plenum outlet, the $[\text{H}_2]\text{FW}$ required for reducing the ECP below the Ecrit at 80% power coastdown was 1.6 ppm, but HWC would not become effective until a 1.8 ppm $[\text{H}_2]\text{FW}$ was reached at 100% power level. The ECP at the upper downcomer outlet would go below the Ecrit when the $[\text{H}_2]\text{FW}$ reached 1.6 ppm at 100% power level, and it would require a 1.5 ppm $[\text{H}_2]\text{FW}$ for the ECP to be effectively reduced at 80% power coastdown. At the bottom lower plenum outlet, the required $[\text{H}_2]\text{FW}$ remained unchanged at 1.4 ppm regardless of the changes in power level. The degree of power coastdown did not result in a consistent trend in the required $[\text{H}_2]\text{FW}$ at the three foregoing locations of Reactor X.

The ECP response to $[\text{H}_2]\text{FW}$ and power level at the recirculation system outlet was comparatively straightforward. The ECP monotonically increased with decreasing power level at Reactor X. While an effective ECP reduction at 0.6 ppm $[\text{H}_2]\text{FW}$ was observed at 100% power, a 0.8 ppm $[\text{H}_2]\text{FW}$ was required at the 80% power level.

Summarizing the predicted ECP results at these four locations, we noted that no significant ECP differences

due to power coastdown were observed when the $[H_2]_{FW}$ was either much less or greater than the critical concentration at which the ECP markedly decreased to below the Ecrit. A 20% coastdown percentage would induce a more oxidizing environment at different locations in a BWR and hence led to a varied HWC efficiency. Furthermore, the impact of a power coastdown on the HWC efficiency would vary from plant to plant. Therefore, a BWR with a different power density and different physical dimensions may experience a different outcome upon power coastdown operations. An individual analysis on the impact of power coastdown on the corrosion mitigation effectiveness of HWC in a BWR is necessary.

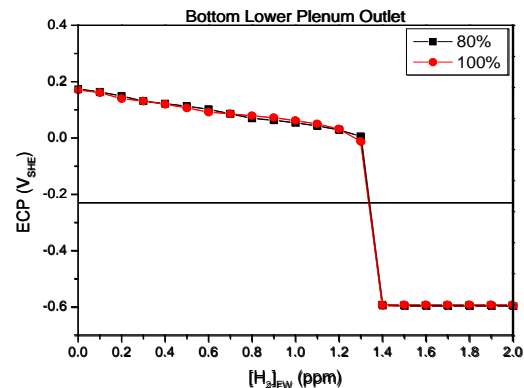
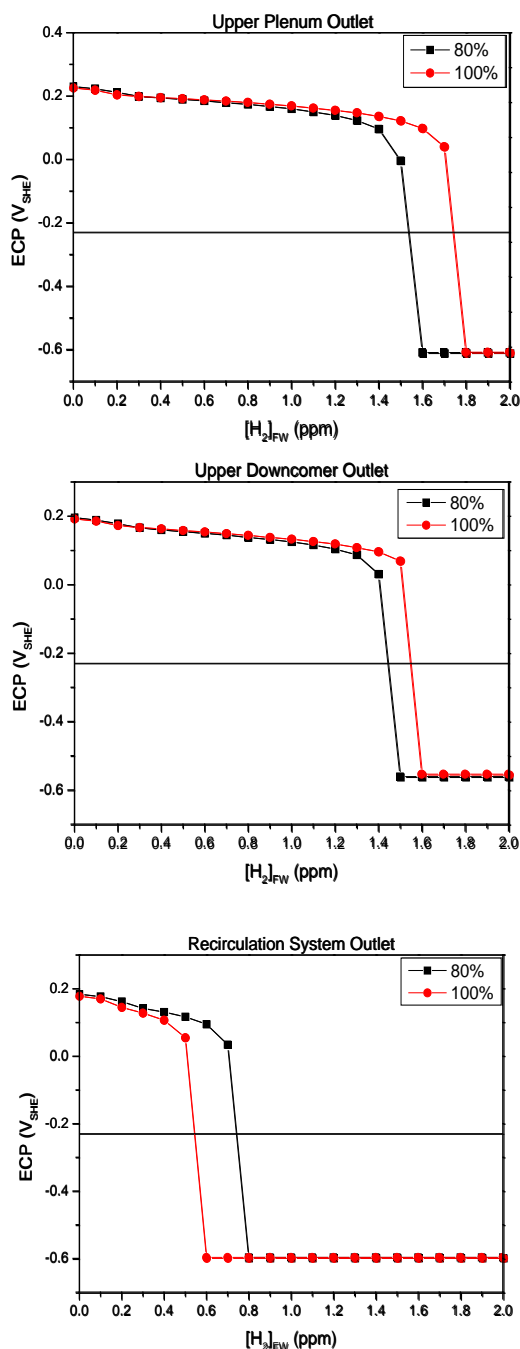


FIG. 6 VARIATIONS IN ECP AS A FUNCTION OF $[H_2]_{FW}$ AT FOUR SELECTED LOCATIONS OF REACTOR X WITH OPERATING POWER LEVELS OF 80% AND 100% OF THE RATED POWER.

Conclusions

The responses of water chemistry and ECP to HWC at some selected locations in Reactor X under postulated power coastdown levels were theoretically evaluated. Upon a power coastdown operation, the degree of radiolysis and coolant residence time in the core of a BWR may vary, causing changes in radiolytic species concentrations and resulting in varied HWC efficiency at different locations.

For the outlets of the upper plenum and the upper downcomer of Reactor X, a 20% power decrease would tend to promote a slightly more oxidizing coolant environment for the structural components at these locations. The $[O_2]$, $[H_2O_2]$, and the ECP monotonically increased with decreasing power level at the recirculation system outlets. In summary, no consistent trend could be found for changes in the effective $[H_2]_{FW}$ as a function of decreased power level in the PCC of Reactor X.

Based upon the predicted results, the impact of power coastdown on the HWC effectiveness in a BWR is expected to vary from location to location and eventually from plant to plant due to different radiation dose rates and physical dimensions. The outcomes derived in this work for Reactor X may not be applicable to other BWRs of the same type, and a new, full-scale analysis is highly recommended.

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